

Some Physics Processes in the Nitrogen-Filled Photoluminescence Cell - Rev. 1

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Some physics processes in the nitrogen-filled photoluminescence cell – Rev. 1

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1. Introduction

As shown in Ref. [1], the photoluminescence cell is a viable candidate for monitoring the total energy in the Linac Coherent Light Source. In Ref. [1], most of the discussion was concentrated on the cell with argon as a working gas. In the present note I provide a discussion of some physics processes that may affect the performance of the photoluminescence cell with the nitrogen fill. In particular, I will consider the role of the space charge effects, ambipolar diffusion, and recombination processes. This group of phenomena determines the duration of the afterglow process that follows an initial short (<100 ns) burst of optical radiation. The presence of this afterglow can be of some significance for the detection system.

Compared to my previous note with the same title UCRL-TR-222274, a more detailed discussion of space charge effects is provided, with an emphasis on the electrostatic confinement of the primary electrons. Also, some additional atomic data are included into sections describing recombination processes.

The general template for this discussion follows a draft report [1] where the argon-filled cell was considered. But some processes in nitrogen are different and require separate consideration. In what follows, I am not attempting to produce "exact" results, but rather to provide a quick order-of-magnitude scoping study.

2. Space charge effects

2.1 Electrostatic potential

To get some orientation in the role of space-charge effects, we consider an electric potential created by a bare ion cylinder of a radius r situated inside a conducting grounded cylinder of a radius R. This very crude model allows us to get some insight into the possible significance of space-charge effects: if the resulting potential well (for the electrons) is much deeper than their characteristic energy, this would mean that the electrons will be bound to the ion core, and we will actually deal with a quasineutral plasma. The potential at the axis of a cylinder of a radius r is

$$\varphi = q(1 + 2\ln\frac{R}{r}),\tag{1}$$

where q is the charge per unit length. For the ions formed initially within the radius of the X-ray beam, r should be identified with the beam radius. The potential difference between the axis and the distance $\sim 2r$ from the axis is

$$\Delta \varphi = q(1 + 2\ln 2) \approx 2.4q \tag{2}$$

2.2 Electrostatic confinement of the primary electrons

The primary electrons will be confined within the radius $\sim 2r$ if their energy is less than $\sim e\Delta\varphi$. The number of the primary ions produced by the X-ray beam per unit length is:

$$N_i^{(p)} \approx n_0 \sigma_{vh} N_X \tag{3}$$

where n_0 is the neutral gas particle density, σ_{ph} is the photoionization cross-section for the incident X-ray beam, N_X is the number of x-ray photons in one pulse, and the superscript "p" stsands for the word "primary". So, the potential well for the primary electrons will be roughly

$$U = e\Delta\varphi \approx 2e^2 n_0 \sigma_{nh} N_x \tag{4}$$

(we rounded the numerical coefficient 2.4 in Eq. (2) to 2, to emphasize the qualitative nature of our analysis). The coefficient $n_0 \sigma_{ph}$ is approximately equal to ε/L , where ε is a fraction of the energy lost by the X-ray beam in the cell, and L is the length of the cell. The number of X-ray quanta is

$$N_X \approx 6.4 \times 10^{12} \frac{Q(mJ)}{E(keV)} \tag{5}$$

In other words, we get the following expression for the electrostatic confinement potential (in "practical" units):

$$U(eV) \approx 1.8 \times 10^6 \frac{A \varepsilon Q(mJ)}{L(cm)E_X(keV)}$$
 (6)

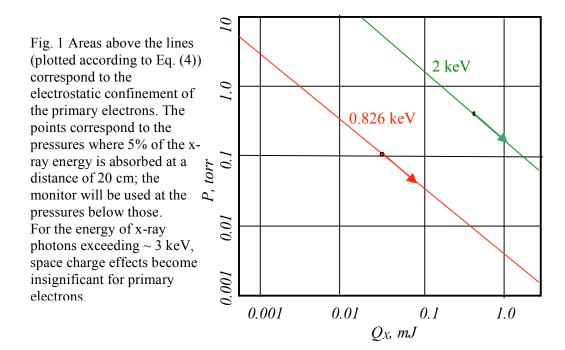
Note that this potential well isolates the primary electrons not only from the side walls but also from the end walls (provided they have the same potential). The condition for the confinement of primary electrons is illustrated in Fig. 1.

If the primary electrons are electrostatically confined, they will pull the ions in the radial direction, causing a gradual expansion of the central column. But this process is relatively slow, especially given the fact that the ions experience a strong friction against the neutral component. The lower-energy electrons produced by the secondary ionizations will be also confined to this narrow zone. So, only in this narrow zone the excited atoms will be formed and the radiation will come out from a very narrow channel near the axis. Given that the radiative life-time of the excited states is in the range of tens

of nanoseconds, the radiation will occur from the zone smaller than a few millimeters in radius

If the energy of the primary electrons exceeds the potential well depth by a factor of 2-3, they will be confined to a radius which is still less than the tube radius R; they will also be confined in the axial direction. The ionization and radiation will be limited to this area. This regime is advantageous in the sense that all the energy of the primary electrons is spent inside the cell volume (no losses of the primary electrons), and that the radiation source is very bright (localized near the axis).

At low gas pressures and at high x-ray energy and/or at a low energy content in the x-ray pulse, the space-charge effects become unimportant for the primary electrons. Their motion under such circumstances is determined by their slowing-down time by ionization and excitation; if this time is long-enough, they can be lost to the walls. We consider this regime for the case where the primary electrons produce the ionization in the whole volume of the photoluminescence cell. We leave refinement related to intermediate regimes for the future work.



2.3 Space charge effects for the secondary electrons

In this section, we consider the situation where the secondary electrons produce a quasi-uniform ionization of the whole volume of the photoluminescence cell. I assume here that the cell is a sphere of a radius R. To see if space-charge effects are significant for the secondary electrons, we again find the depth of the potential well formed by the uniformly distributed bare ion charge (one, of course, could also use the Debye radius arguments, which are equivalent to our present approach). The depth of the electron potential well at the center of the sphere is now

$$U = \frac{e^2 N_i}{2R} \tag{7}$$

where N_i is the total number of ions produced in the cell (see next section). The secondary electrons will be well confined by the space charge idf their characteristic energy is lower than U by a factor \sim 5-10. We will return to this issue in one of the subsequent sections.

3. The total number of ionizations.

If the primary photoelectron loses its energy before hitting the wall (as is the case in the regime of the electrostatic confinement), the total number of ionizations is

$$N_i \approx A n_0 \sigma_{nh} N_{\chi} L \tag{8}$$

where A (=50 – 200) is the number of ions produced by one primary electron during its deceleration to the energy below the ionization potential, and L=2R is the path length of the X-ray beam in a gas. The parameter A does not depend on the gas density. Therefore, the total number of ions (and optical photons) in this case if proportional to the gas pressure.

We make a rough approximation

$$\frac{A}{E_X(keV)} \sim 50 \tag{9}$$

In the regime of electrostatic confinement of the primary electrons, Eqs. (5), (8), and (9) lead to the following "practical" expression for N_i :

$$N_i = 3.2 \times 10^{14} \varepsilon Q(mJ) \tag{10}$$

One can see that this number is small compared to the total number of neutral atoms in the ionized region, i.e., the gas remains weakly ionized.

For the case where there is no electrostatic confinement and the primary electrons hit the wall before losing their energy, only a fraction ε_i <1 of this number of ions is created, $\varepsilon_i \sim R/l$, where l is the distance at which the primary electron finds itself from its birth point after losing its energy. It is roughly equal to the electron range and is inversely proportional to the gas density,

$$l = \frac{1}{n_0 \sigma_{eff}} \tag{11}$$

where σ_{eff} is some effective cross-section that one can introduce to characterize this process. In this regime we obtain for the total number of ions

$$N_i \approx 2A\sigma_{ph}\sigma_{eff}n_0^2R^2N_X \tag{12}$$

Detailed information regarding the electron range can be found in Ref. [2] provided by S. Hau-Riege.

We see that the total number of ions (and total number of optical photons) is proportional to the square of the gas pressure. Eq. (11) can be rewritten in the following way:

$$N_i \approx A \varepsilon \varepsilon_i N_X$$
, (13)

or, accounting for (5) and (9),

$$N_i = 3.2 \times 10^{14} \varepsilon \varepsilon_i Q(mJ) \tag{14}$$

4. Electrostatic effects for the secondary electrons

With this result in mind, Eq. (7) can be presented as:

$$U(eV) = 2.3 \times 10^7 \frac{\varepsilon \varepsilon_i Q(mJ)}{R(cm)}$$
 (15)

Taking Q=2 mJ, $\varepsilon=0.01$, $\varepsilon_i=0.1$ and R=10 cm cm, we find from Eqs. (5) and (6) that $U=4.6\cdot10^3$ eV. In other words, space charge effects for the secondary electrons are very significant and would prevent them from leaving the ionized core. The temperature of the secondary electrons will end up at the level of a few electron-volts (it must be well below the excitation threshold).

In other words, in the final state we get a weakly ionized plasma with the electron temperature of a few electron-volts. This state is reached within the time of order of 10 or so nanoseconds (depending on the gas density). The lifetime of typical excited states is of order of a few tens of nanoseconds. So, the first burst of optical radiation comes out within the time shorter than 100 ns.

For a spherical cell of a radius R, the ion density will be

$$n_i = \frac{3N_i}{4\pi R^3},\tag{16}$$

or, using Eq. (7),

$$n_i(cm^{-3}) = 0.75 \times 10^{14} \frac{\varepsilon \varepsilon_i Q(mJ)}{[R(cm)]^3}.$$
 (17)

This density is orders of magnitude less than the neutral gas density.

Perhaps, introduction of a very weak magnetic field ($\sim 150~G$ for the X-ray energies up to 2. 5 keV) could make the performance of the cell more predictable, by localizing the energy deposition by the primary electrons (the gyro-radius of 2.5 keV electrons in a 150 G field is roughly 1.2 cm).

4. Energy equilibration with the neutral gas

The collisions with the gas molecules will gradually cool the electrons down. This takes time of roughly

$$\tau_{cooling} = \frac{M}{m} \frac{1}{n_0 \sigma_{e0} V_{Te}} \quad , \tag{18}$$

where m and M are the electron and the molecule mass, respectively, and σ_{e0} is the electron-molecule cross-section. Taking as a representative value for the latter 10^{-15} cm², we find that

$$\tau_{cool}(s) \sim \frac{2.4 \times 10^{-5}}{p(torr)\sqrt{T(eV)}}.$$
(19)

Taking $T \sim 1/40$ eV and p ~ 1 torr, one finds that the temperature equilibrium will be reached within ~ 0.1 ms.

The ion component of this plasma will consist of molecular ions, N_2^+ , and atomic ions N^+ ; we neglect a small admixture of N_2^{++} . The ratio of the densities of N_2^+ and N^+ is approximately 3 [3,4]. The composition may be important in the evaluation of the dissociative recombination. The ion temperature stays close to the gas temperature during all the times of interest for us.

5. Volumetric recombination

For the molecular ions, the recombination process will be dominated by the dissociative recombination. According to Refs. [5,6],

$$\tau_2(s) = \frac{3 \times 10^6}{n_i (cm^{-3})}. (20)$$

For the ion density of 10^8 cm⁻³, this time is ~ 0.03 s. The atomic ions will recombine via radiative recombination, which is slow [7]. The 3-body recombination recombination time at room temperature, (1/40) eV, with the 3^{rd} body being another electron, is:

$$\tau_3(s) = \frac{7 \times 10^{18}}{\left[n_i \left(cm^{-3}\right)\right]^2}.$$
 (21)

For the ion density of 10^8 cm⁻³ (see Eq. (17)), this time is orders of magnitude longer then the distance between the pulses.

6. Ambipolar diffusion

As the volumetric recombination is quite slow, the ionized state will decay by the diffusion of the ions to the walls and the surface recombination. This diffusion will occur at a rate determined by the ion mean-free path λ_i determined by elastic scattering on the neutrals and charge exchange process. As a representative value of this mean free path we take the following estimate:

$$\lambda_i(cm) \sim \frac{10^{-2}}{p(torr)} \tag{22}$$

The diffusion coefficient at room temperature will then be (for nitrogen)

$$D(cm^2/s) \sim \frac{200}{p(torr)} \tag{23}$$

For a sphere of a radius R, the diffusion time to the walls can be evaluated as

$$t_{diff} \sim \frac{R^2}{6D} \tag{24}$$

or, in "practical" units,

$$t_{diff}(s) \sim 10^{-3} [R(cm)]^2 p(torr)$$
 (25)

For the expected range of pressures in the gas cell, the diffusion time is typically longer than the distance between the successive pulses at 120 Hz rep rate. This means that there will be a continuous presence of the ionized component in the cell (unless the recombination is fast enough). The time of the gas replacement in the cell due to the continuous outflow through the 3-mm diameter end apertures for the assumed radius of the cell R~ 10 cm is of the order of 1 s, much longer than 1/120 s separation between the pulses.

Summary

The first short burst of the optical radiation will be followed by a much longer afterglow. The duration of the latter, for the typical parameters, will be determined by the ambipolar diffusion to the walls, with a subsequent surface recombination, and by the gas outflow through the end apertures.

Some effect on the conclusions of this paper may have the formation of the negative molecular ions N_2 . The negative atomic ions N do not exist. I did not have enough time to find out whether the molecular ions exist.

A universal cure for reducing the afterglow time (be it needed) is introduction of a weak guiding magnetic field of ~ 150 G. This field will lead to a reduction of the radius of the ionized zone, increase of the ion density, and corresponding increase in the recombination rate. It will also reduce the losses of the primary ions to the wall and improve the optical yield at lower pressures/lower pulse energy contents, where the primary electrons are not confined by the space charge.

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References.

- 1. D. Ryutov, R. Bionta, and S. Hau-Riege. "Photoluminescence for non-destructive total energy measurements and imaging of X-ray beam in the Linac Coherent Light Source," UCRL-TR-220017-DRAFT, March 2006.
- 2. J.L. Barett and P.B. Hays. J. Chem. Phys., 64, 743 (1976).
- 3. E. Krishnakumar and S.K. Srivastava. "Cross sections for the production of N_2^+ , $N^++N_2^{++}$ and N^{++} by electron impact on N_2 ", J.Phys. B: At. Mol. Opt. Phys., **23**, 1893 (1990).
- 4. H.C. Straub, P. Renault, B.G. Lindsay, K.A. Smith, R.F. Stebbings. "Absolute partial cross sections for electron-impact ionization of H₂, N₂, and O₂ from threshold to 1000eV," Phys. Rev. A, **54**, 2146 (1996).
- 5. H. Massey. "Aomic and molecular collisions." Taylor and Francis Ltd, London, 1979, p. 123.
- 6. J.B. Hasted. "Physics of atomic collisions," Butterworths, London, 1972, p. 423.
- 7. D.L. Book, NRL Plasma Formulary, NRL Publication 0084-4040, 1987.